Metal-Insulator Transition of the Ba(3×1) Linear Chain Reconstruction

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INTRODUCTION

The study of adatom structures on surfaces such as the Si(111) surface can serve to develop an understanding of model sytems in one and two dimensions, which bear high relevance for both an understanding of physics in low dimensions and applications such as ultra-small scale microelectronic structures. At this scale, atomistic bonding configurations play an important role, and local charge distributions can critically affect the energy and dispersion of an adatom band. Local correlation effects, usually characterized by a Hubbard U potential, determine wether or not the Fermi level will be set within a band and thus decide between metallic or insulating behavior of the adatom system.

Of particular interest is the class of (3×1) reconstructions formed on Si(111) by alkali, alkali-earth and silver atoms. It consists of one-dimensional chains of atoms. While the alkali atoms are found to form a true semiconducting system [1], simple electron counting arguments lead to the assumption that an alkali-earth (3×1) reconstruction would be metallic. The first published study of such kind was the work on Mg(3×1) by An et al. [2] using photoelectron spectroscopy; the system is in fact found to be insulating. In this work we have chosen the Ba(3×1) reconstruction to study the effects of additional or lacking charge in the chain because the large Ba atom radius should ensure sufficient overlap of the charge clouds between two metal atoms. Various structural models are discussed in the literature, but recently refined calculations seem to favor a chain-channel reconstruction [3,4], as is schematically shown in Fig.1.

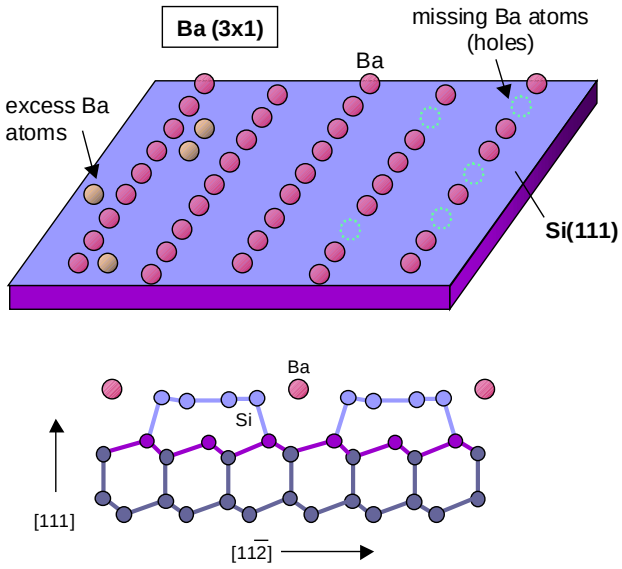

Figure 1:
Schematic of the Ba(3×1)/Si(111) reconstruction. It is possible to prepare the system with excess or missing Ba atoms (top). The chain-channel model is currently discussed in the literature as the most likely one. Removal of Ba atoms does not involve bond breaking and leaves a hole-doped one-dimensional chain (bottom).

EXPERIMENT

The ARPES experiments were carried out at beamline 7.0.1 at the Advanced Light Source. The energy resolution used was ~60 meV, the angular resolution was ~1/4º. A vicinal Si(111) substrate with 2º miscut was used to guide formation of a single-domain reconstruction. A monolayer of Ba was evaporated onto a Si(111)-(7×7) surface from a commercial getter source and annealed in front of a LEED screen at ~600 ºC until a sharp (3×1) reconstruction was obtained.

RESULTS AND DISCUSSION

The quality of the preparation of the Ba(3×1) reconstruction was tested and confirmed by its LEED pattern, shown in Fig. 2, which reveals the one-dimensional structure of the adatom chains.

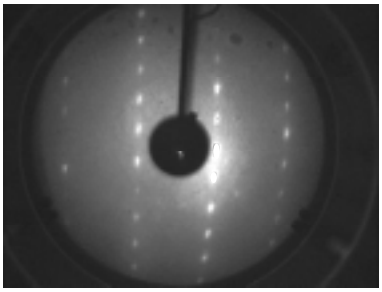

Figure 2:
LEED image of the single-domain (3×1) reconstruction, recorded at 84 eV.

Overview valence band maps, taken by photoemission over a wide angular range and with binding energies covering the whole width of the valence band, do not reveal any Fermi level crossing under standard preparation conditions, i.e. the system is found to be insulating. None of the bands shows any features that would have a symmetry point half-way towards the Brillouin zone boundary. Also, no such "3×2" symmetry is seen in the LEED images down to very low electron energies. Therefore we believe it is safe to assume that the system is not in a Peierls-distorted state at room temperature.

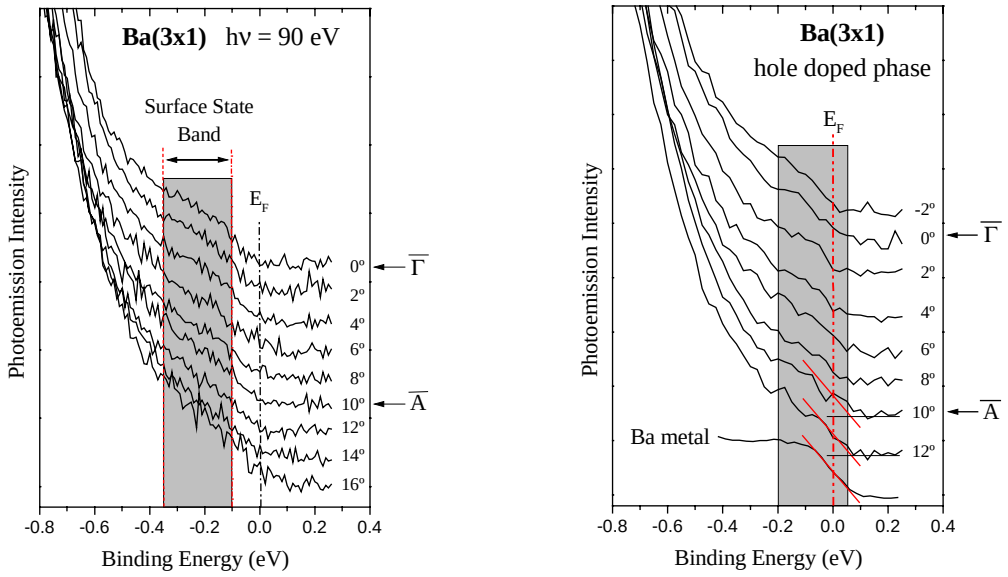

Figure 3:
The left panel shows the surface state band of the Ba(3×1) recontruction under typical preparation conditions. A weakly dispersing band is identified around ~0.3 eV below E_F. Upon sufficient Ba desorption (right panel), the band is shifted upward by about 0.2 eV such that the band crosses E_F.

We have taken close-up band maps in the vicinity of the Fermi level to inspect any bands close to E_F in more detail. We find a band extending over a binding energy range of 0.35 eV to ~ 0.10 eV below E_F, with very weakly dispersing features (Fig. 3, left panel). When looking at the data as presented in Fig. 3, one must be aware of the very low photoemission cross-section of the Ba states at the photon energy used, an effect which gains further impact because of the low coverage (~1/3 monolayer).

In an attempt to modify the amount of charge contained in the topmost band – and hence to induce a metallic condition - we have added and removed atoms from the surface by additional Ba evaporation or desorption, respectively. A surplus of Ba atoms does not render the system metallic, unless large excess amounts of the metal lead to a disordered metallic overlayer. This was previously observed for the Na(3×1) reconstruction [5]. Concerning depletion experiments, an important finding is that an intact (3×1) structure can be prepared Ba-deficient by thermal desorption. Under this condition, when inducing a loss of ~ 10–20 % of the Ba atoms as judged from the Ba 4d core levels, the topmost band moves upward by about ~ 0.2 eV. A Fermi level crossing can then be identified in the vicinity of the zone boundary at $\overline{A}$ (Fig. 3, right panel). The shifting of the band is found to be more pronounced upon further desorption of Ba atoms. From a LEED study, the reconstruction is found to remain intact if up to ~1/4 of the Ba is desorbed. Higher depletion leads to the development of rotated domains, and ultimately to the destruction of the reconstruction.

CONCLUSION

The observed behavior appears consistent with an interpretation as hole-doping of the insulating metal adatom chains. The band in the normal insulating state is only weakly dispersing – indicative of a high degree of localization and the importance of the local charge environment that surrounds the metal adatom. This makes it plausible that the system is a Mott insulator that experiences a local Hubbard U which separates bonding and antibonding orbitals. The energetics of the charge distribution which will also involve the substrate apparently favors a fully filled lower Hubbard band. The shift upward in the Ba-deficient chain then indicates that the highest occupied band has been depleted of electronic charge, and the Fermi level moves into that band, thus reflecting the transition to the metallic state.

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[1] K. D. Lee, J. Chung, Phys. Rev. B. **57**, 2053 (1998).
[2] K. S. An, R. J. Park, J. S. Kim et al., Surf. Sci. **337**, L789 (1995).
[3] S. Erwin, H. Weitering, Phys. Rev. Lett. **81**, 2296 (1998).
[4] A. A. Saranin, A. V. Zotov, V. G. Lifshits et al., Surf. Sci. **426**, 298 (1999).
[5] J. J. Paggel, G. Neuhold, H. Haak, K. Horn, Surf. Sci. **414**, 221 (1999).